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Survey of Postirradiation Heat Treatment as a Means to Mitigate Radiation Embrittlement of Reactor Vessel Steels

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Thermostructural Materials Branch
Material Science and Technology Division

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Postirradiation heat treatment (annealing) is being investigated as a method for the alleviation of radiation embrittlement to reactor vessel steels. Objectives of this study were to identify those service and metallurgical variables which can affect annealing response, to report data comparisons available for qualification of suspected influences, and to survey experimental results from commercially produced reactor materials and duplicate materials from the laboratory.

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20. Abstract (Continued)

Twelve factors are identified as having potential for affecting the postirradiation heat treatment response of low-alloy pressure vessel steels. A tentative qualification of six factors as significant contributing variables is made on the basis of experimental comparisons. Of these, heat-treatment temperature was observed to exert the largest influence on the embrittlement relief by annealing. The evidence indicates that a 399°C heat treatment, but not a 343°C heat treatment, is a practical and effective method for reducing and controlling radiation embrittlement in reactor vessels.

CONTENTS

SURVEY OF POSTIRRADIATION HEAT TREATMENT AS A MEANS TO MITIGATE RADIATION EMBRITTLEMENT OF REACTOR VESSEL STEELS

INTRODUCTION

Nuclear-radiation service typically produces a progressive reduction in the notch ductility of low-alloy steels. The reduction is manifested by a decrease in Charpy-V (C_v) upper-shelf energy level and by an elevation in temperature of the ductile-to-brittle transition. Postirradiation heat treatment (annealing) is being investigated as a method for the reversal of these detrimental radiation effects for reactor-vessel steels. This study was undertaken to analyze factors which could affect annealing response, report data available to qualify suspected influences on annealing, and summarize experimental results generated for many commercially produced reactor materials and companion materials produced in the laboratory.

The degradation of notch ductility by radiation is recognized by the ASME Code (Section III) [1] and by the Code of Federal Regulations (10CFR50) [2], and direct and indirect limitations are imposed on postirradiation properties. For example a C_v upper-shelf energy of at least 68 J (50 ft-lb) is required by the ASME Code by its use of this energy level to index the reference nil-ductility temperature, RT_{NDT}, for fracture toughness characterization. If notch ductility properties fall below Code requirements, application of an in-service heat treatment is one option open for extending vessel life. That is, the Code of Federal Regulations states: "Reactor vessels for which the predicted value of adjusted reference temperature exceeds 96°C (200°F) shall be designed to permit a thermal annealing treatment to recover material toughness properties." An alternate option involves performing an essentially complete volumetric inspection of the beltline region of the vessel and development of a fracture mechanics analysis which "conservatively demonstrates, making appropriate allowances for all uncertainties, the existence of adequate margins for continued operations" [2].

At present, there is little doubt that Code notch ductility requirements can be met by recently constructed reactor vessels over projected service lifetimes. A concern does exist, however, for certain vessels produced prior to 1971 which did not have the benefit of current radiation effects technology. Here, a combination of factors including high copper and phosphorus contents and low initial C_v upper-shelf energy level may exist which could bar meeting the Code criteria after some period of vessel operation. A high content of copper and phosphorus impurities in steel is known to contribute a high sensitivity to radiation-induced reductions in notch ductility. A low upper-shelf energy, in addition, provides less of a margin for radiation-induced degradation. NRC Regulatory Guide 1.99 [3] is available for predicting radiation effects to reactor vessel steels from knowledge of the steel composition and the expected neutron-fluence accumulation by the vessel.

Manuscript submitted October 10, 1978.

Existing data on recovery of notch ductility with postirradiation heat treatment are quite limited and are largely in the form of C_v data. For the most part those data developed prior to 1972 represent only spot tests of annealing response and thus do not fully describe upper-shelf and transition behavior. Increased interest in the annealing method since 1972 has led to specific studies of annealing behavior. More importantly, current studies are including compact specimens (CS) for fracture toughness (K_{Ic}, K_{Id}, K_{Jd}) determinations [4-7]. Initial CS results from U.S. programs are expected in 1979 and 1980. To guide the investigations, important data trends have been discerned from the existing C_v data, as reported below.

A discussion of vessel annealing operations is beyond the scope of this report; however, two annealing options appear to be available to reactors. Heat treatments at temperatures up to 343°C (650°F) represent one option, with primary-pump or nuclear heating being used to attain the requisite temperature in the vessel. With this option the reactor coolant and the core internals would be left in place. This annealing option has already been successfully used for a pressurized water reactor (Army SM-1A reactor) [8, 9]. In this case, however, the vessel operating temperature of 221°C (430°F) was significantly lower than that of commercial power reactors, 288°C (550°F).

The second option involves the use of auxiliary heaters to bring the vessel (or selected components thereof) to a higher annealing temperature, such as 399°C (750°F). Higher temperatures can result in greater recovery of properties, but removal of the core internals as well as the coolant would be required. For most reactors the mechanics of annealing at temperatures significantly above 399°C (750°F) or 427°C (800°F) would appear to be prohibitive.

POTENTIAL FACTORS INFLUENCING HEAT-TREATMENT RESPONSE

Several variables are considered to have potential for influencing notch ductility recovery by postirradiation heat treatment. By category these include

<u>Service Variables</u>

Irradiation temperature,

Neutron fluence,

Relative radiation resistance,

Applied stress,

Steel Metallurgy

Impurity element composition (including Cu, P, S),

Alloy element composition,

Product form (plate, weld, or forging),

Heat Treatment

Annealing temperature,

Annealing time,

Cyclic Irradiation and Annealing Conditions

Annealing time and temperature,

Fluence before first anneal, and

Fluence between anneals.

Synergistic effects between variables are also considered possible.

PRESENT QUALIFICATION OF SUSPECT VARIABLES

Experimental data are available from 288°C radiation experiments (accelerated-test reactor irradiations) to directly or indirectly test the contribution of some suspect variables. It is emphasized that the observations are tentative in some cases and require confirmatory tests.

Irradiation Temperature

Notch ductility recovery with 343 to 427°C postirradiation heat treatments appears to increase with decreasing irradiation temperature in the range of 343 to 121°C. Experimental results in support of this indication are given in Figs. 1 through 6. The figures index the irradiation and annealing effect to the conventional (arbitrary) C_v 41-J (30 ft-lb) transition temperature; however, transition-temperature changes measured at C_v 41 J and C_v 68-J energy indices show good agreement in most cases. Figures 1 to 5 show results for the ASTM A302-B reference plate [10]. Figure 6 shows results for two A533-B Class 1 plates from the same (split) steel melt [12]. (The data of Figs. 1 and 2 and of Figs. 3 and 4 were developed using one irradiation assembly with two-zone temperature control. Likewise, the data of Fig. 6 were developed with a single irradiation experiment.)

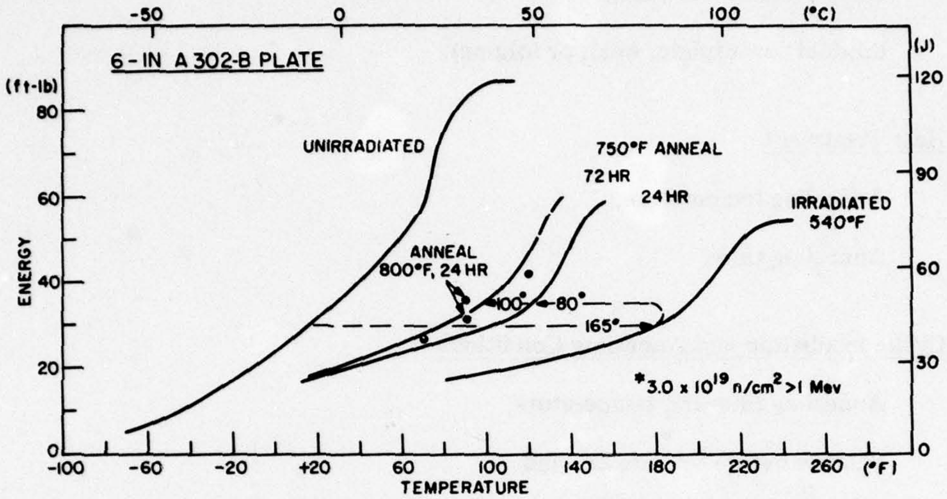

Fig. 1 — Response of the ASTM A302-B reference plate to various low-temperature heat treatments after 282°C (540°F) irradiation (experiment A). (From Ref. 10.)

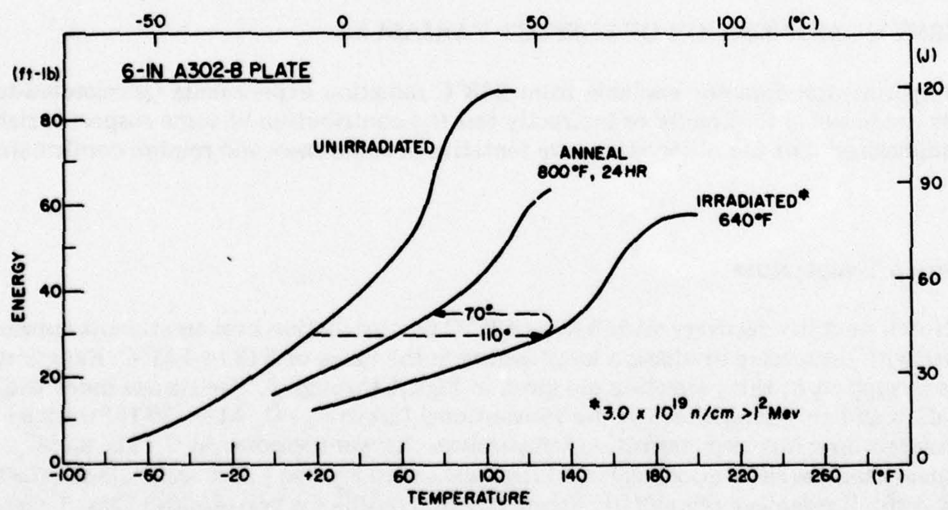

Fig. 2 — Response of the ASTM A302-B reference plate to a 427°C (800°F) heat treatment after 338°C (640°F) irradiation (experiment A; see Fig. 1 also)

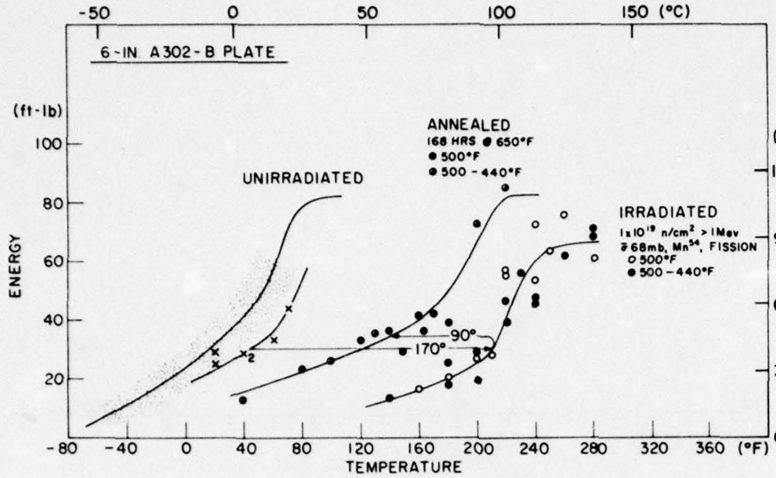

Fig. 3 — Charpy-V-notch ductility behavior of the ASTM A302-B reference plate before and after irradiation at a constant temperature of 260°C (500°F) and under cyclic temperature conditions ranging from 260 to 227°C (500 to 440°F). Responses of both steel conditions to 343°C (650°F) postirradiation annealing for 168 hr are also shown. The x data points depict the unirradiated behavior of the particular section of A302-B steel used for this experiment (experiment B). (From Ref. 11.)

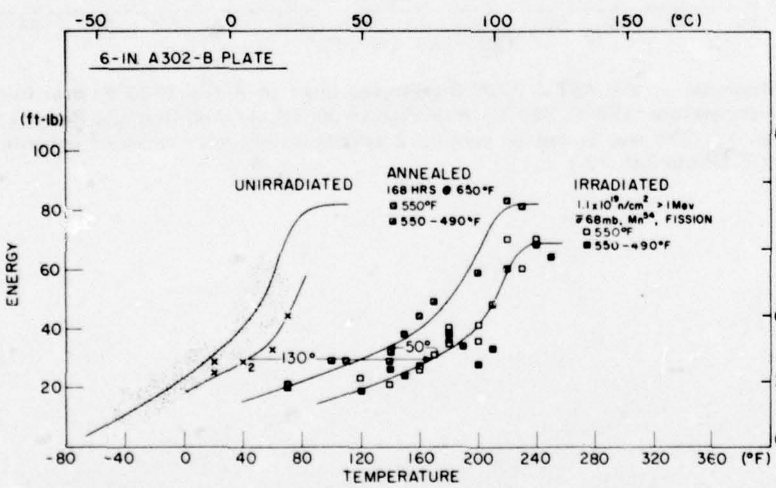

Fig. 4 — Charpy-V-notch ductility behavior of the ASTM A302-B reference plate before and after irradiation at a constant 288°C (550°F) and under cyclic temperature conditions ranging from 288 to 254°C (550 to 490°F). Responses of both steel conditions to 343°C (650°F) postirradiation annealing for 168 hr are also shown. The x data points depict the unirradiated behavior of the particular section of A302-B steel used for this experiment (experiment B; see Fig. 3 also). (From Ref. 11.)

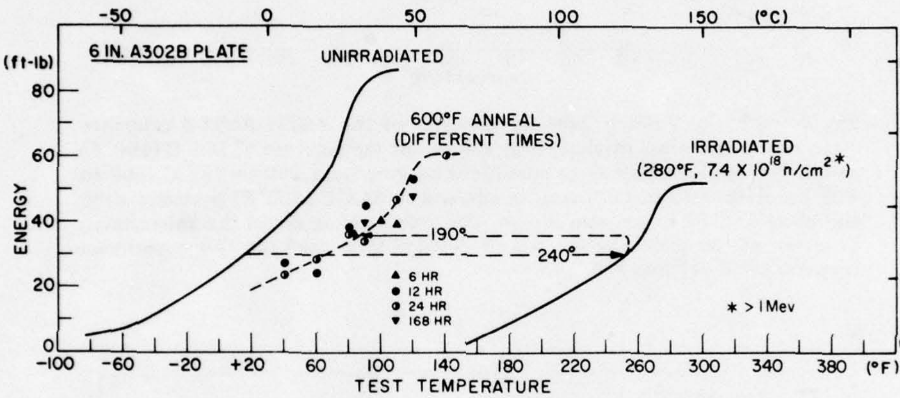

Fig. 5 — Response of the ASTM A302-B reference plate to 316°C (600°F) heat treatment after low-temperature (138°C, 280°F) irradiation in the Brookhaven Graphite Reactor (BGR) (experiment C). The results can be compared approximately with those of experiment B, Figs. 3 and 4. (From Ref. 10.)

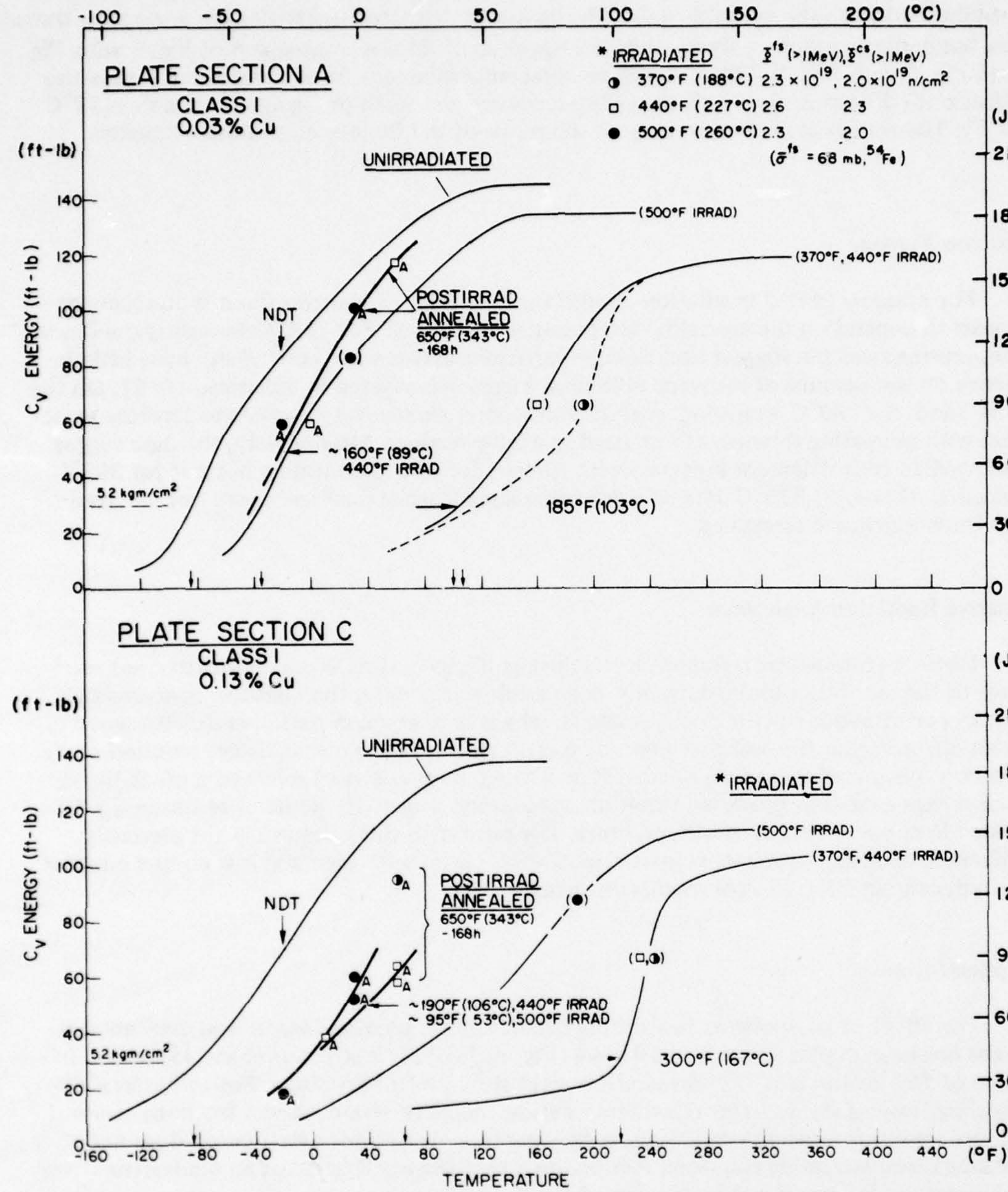

Fig. 6 — Charpy-V-notch ductility recovery in two plates from a split melt of A533-B steel (different copper contents) with 343°C (650°F) 168-hr heat treatment following irradiation at different temperatures. Greater residual embrittlement after heat treatment is observed for the plate with the higher copper content (plate section C). (From Ref. 12.)

The effect of irradiation temperature on residual embrittlement after annealing cannot be established from the available data. The data are too limited and typically concern the transition-temperature regime only and not the upper shelf. In the comparison of Fig. 1 with Fig. 2 and Fig. 3 with Fig. 4, differences in residual embrittlement from transition temperature indications (ΔT irradiation—ΔT annealing recovery) are small or negligible: less than 17°C (30°F). The similarity, however, may be the result of the fluence conditions evaluated.

Neutron Fluence

For nominal 288°C irradiation conditions the effect of neutron fluence on recovery appears to depend on the annealing temperature employed. For 343°C annealing the limited comparisons available suggest that fluence variations above $\approx 1 \times 10^{19}$ n/cm^2 have little influence on the amount of recovery obtained. Figure 7 is offered in illustration [13]. On the other hand, for 399°C annealing, transition-temperature recovery appears to increase somewhat with increasing fluence, as discussed in a later section. Alternatively, the data suggest that residual embrittlement increases with fluence for 343°C annealing but not for 399°C annealing. However, 399°C data sets reflecting large fluence differences are not yet available for a conclusive assessment.

Relative Radiation Resistance

Distinct from neutron fluence level, the significance of radiation embrittlement resistance to the recovery obtained has not been established. Here, the question concerns the relative performance of two steels, A and B, which have equal radiation embrittlement but which differ in that the embrittlement of steel A (high radiation sensitivity) required a low-fluence exposure whereas that of steel B (intermediate sensitivity) required a much higher fluence exposure. The proposed variation in radiation sensitivity could arise through a difference in copper content or other factors. The answer to this question is not presently available; however, experiments involving A302-B plates with high and low copper content are underway at NRL to explore this uncertainty [14].

Applied Stress

The effect of an applied stress during irradiation on postirradiation heat-treatment response has been explored for A302-B steel (Fig. 8) [15]. The stress level employed was in excess of 75% of the 288°C preirradiation yield strength (0.2% offset). Following 399°C annealing, essentially full transition-temperature recovery was observed for both stressed and unstressed (reference) conditions, suggesting that stress during irradiation does not alter postirradiation annealing response. It is believed that the application of an equivalent stress during the anneal would not have affected the outcome.

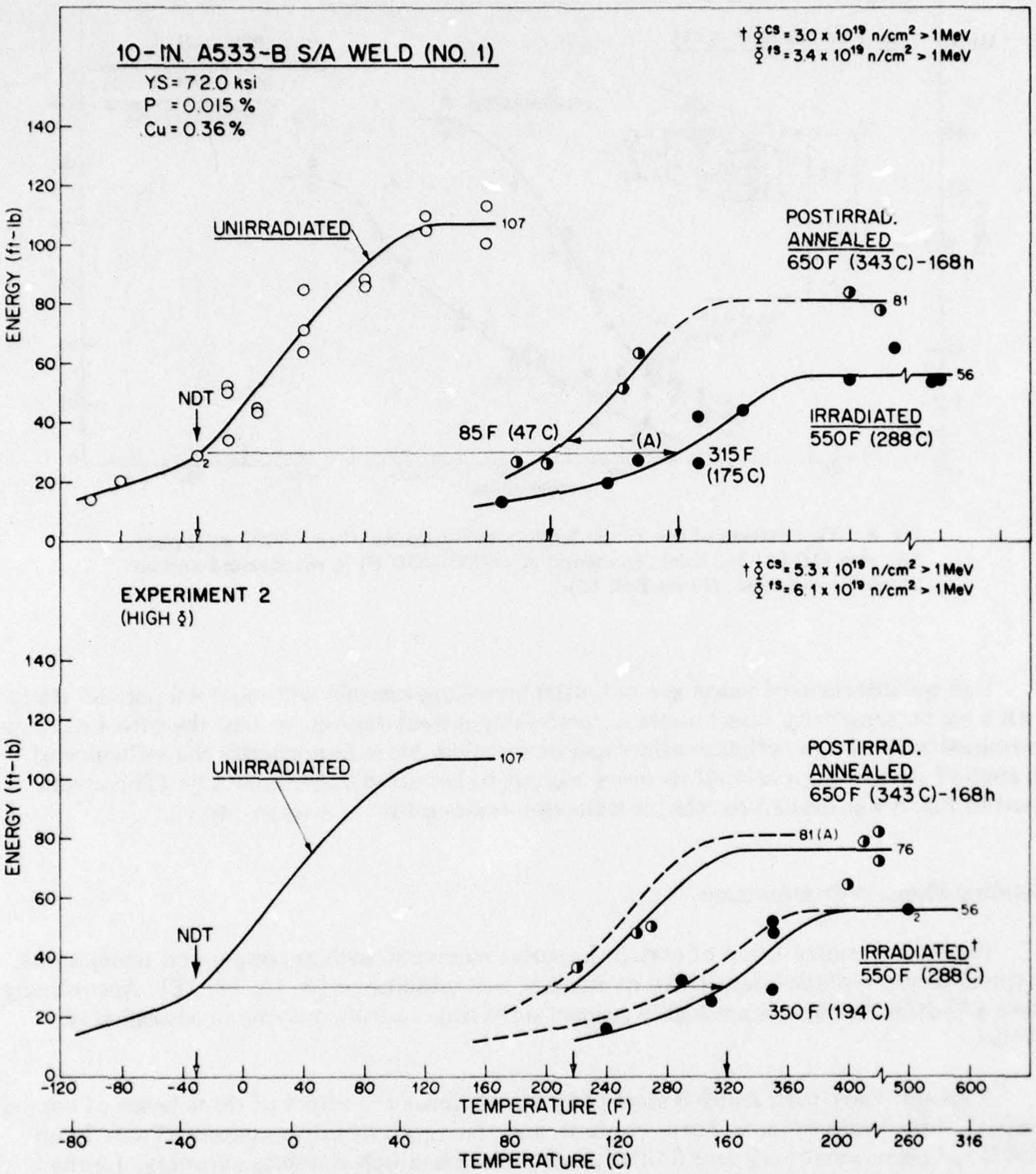

Fig. 7 — Notch ductility of weld 1 (series 1) before and after irradiation to two fluence levels. Notch ductility recovery with 343°C (650°F) 168-hr heat treatment is also shown. (From Ref. 13.)

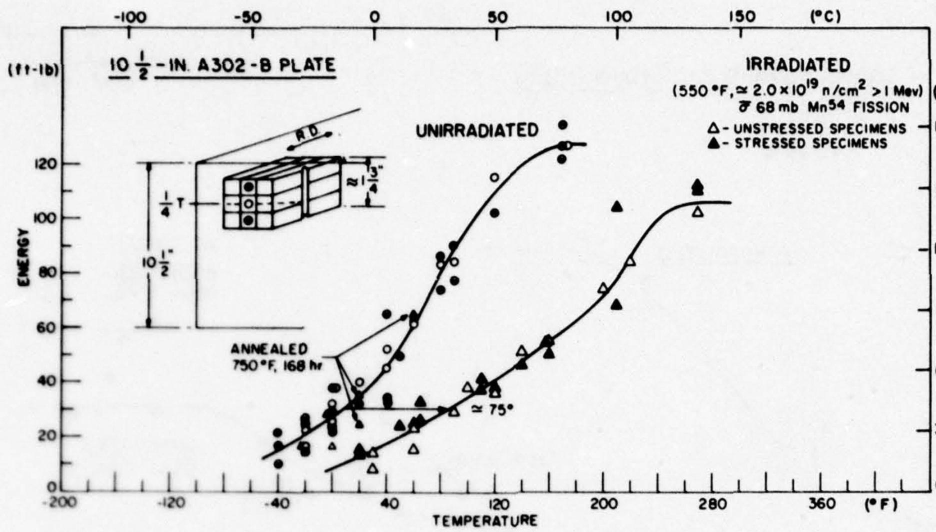

Fig. 8 — Comparison of the notch ductility performance of an A302B steel plate, 267 mm (10-1/2 in.) thick, irradiated at 288°C (550°F) in the stressed and unstressed conditions. (From Ref. 15).

Full qualification of stress as a potential annealing variable will require a parallel study with a higher sensitivity vessel material, preferably a weld deposit, so that the effect of stress combined with a large radiation effect can be revealed. More importantly the influence of an applied stress on upper-shelf recovery has yet to be tested experimentally. (The assessment in Fig. 8 was limited to relative transition-temperature recovery only.)

Residual Element Composition

The strong contribution of certain impurity elements, such as copper and phosphorus, to the observed radiation sensitivity of steels is well established [3, 12, 13, 16]. Accordingly these and other impurities are highly suspect of having a significant role in annealing response.

A recent study with A302-B steel [14] has explored the effect of three levels of copper content, three levels of phosphorus content, and two levels of sulfur content (Table 1) on 288°C radiation sensitivity and 343°C postirradiation notch ductility recovery. Limited tests of the 343°C annealed condition were also conducted. Figures 9, 10, and 11 show the findings on transition-temperature change for the test plates. Full upper-shelf recovery with 343°C annealing was reported for all but one of the plates (composition V67).

Table 1 — Chemical Composition of A302-B Plates

Plate Code	Melt No.	Cast No.	Chemical Composition (wt-%)[a]								
			S	P	Cu	C	Mn	Si	Mo	Al[b]	N
V61	1	1	0.017	0.002	0.15	0.23	1.38	0.26	0.50	0.028	0.001
V63	1	2	0.017	0.014	0.15	0.24	1.28	0.26	0.49	0.029	0.002
V65	1	3	0.017	0.024	0.16	0.24	1.36	0.26	0.50	0.029	0.001
V67	1	4	0.017	0.024	0.29	0.24	1.37	0.26	0.50	0.028	0.001
V71	2	1	0.029	0.016	0.045	0.25	1.23	0.25	0.49	0.027	0.001
V73	2	2	0.029	0.016	0.16	0.25	1.23	0.25	0.49	0.027	0.001
V75	2	3	0.029	0.016	0.30	0.25	1.23	0.25	0.49	0.027	0.001
V77	2	4	0.029	0.024	0.30	0.25	1.23	0.25	0.49	0.027	0.001
A302-B Specification			0.040 max.	0.035 max.	—	0.25 max.	1.15 / 1.50	0.15 / 0.30	0.45 / 0.60	—	—

[a]Ladle analyses by U.S. Steel Corporation.
[b]Total aluminum.

Several experimental determinations were accomplished by the study. First, the effectiveness of 343°C annealing was shown to be relatively independent of copper, phosphorus, or sulfur content for the composition range investigated. Second, the absolute transition-temperature recoveries of the test plates were found to be relatively independent of the magnitude of the prior transition-temperature elevation by irradiation. This observation indirectly suggests that absolute recovery with 343°C annealing is also relatively independent of fluence level. (In Figs. 9 and 10 overall differences in transition temperature elevation approach a factor of 2.) With higher temperature annealing (Fig. 11) a comparable independence of recovery on prior radiation embrittlement was noted. One exception was found in plate V63, for which an explanation could not be offered. A third observation was that a high (0.30%) copper content is detrimental to upper-shelf recovery (343°C annealing) when the steel has a sulfur content on the order of 0.017% S. In contrast, phosphorus was found to have neither a positive or negative effect on recovery for the compositions investigated.

This recent study clearly provides valuable new information on impurity element contributions and annealing trends. The findings however require confirmation for other types of vessel steels and weld metals. In particular, the role of copper and phosphorus in upper-shelf recovery of weld metals requires more detailed study.

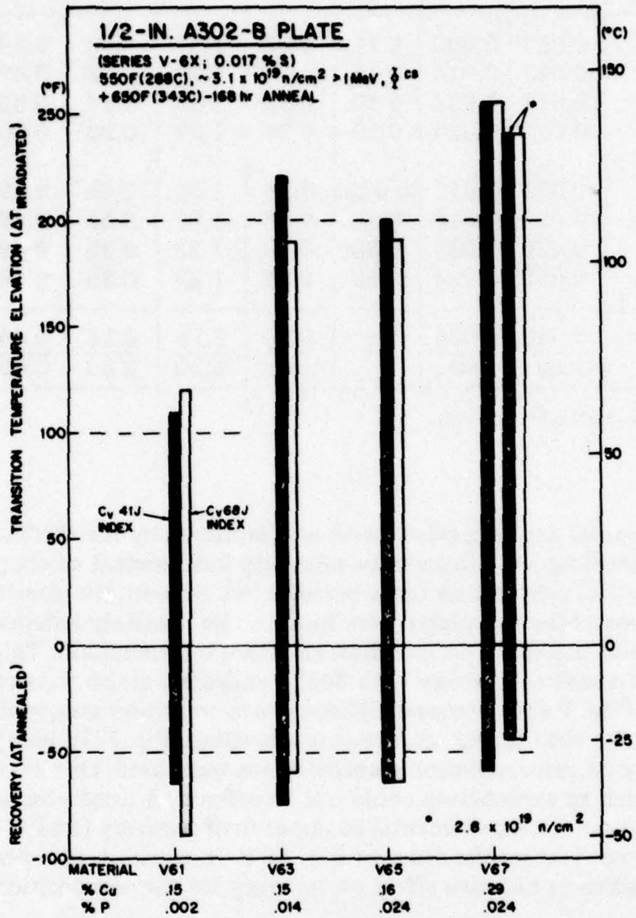

Fig. 9 — Summary of Charpy-V transition-temperature changes (41-J and 68-J indices) of plate series V6X with 288°C (550°F) irradiation and with 343°C (650°F) postirradiation heat treatment. (From Ref. 14.)

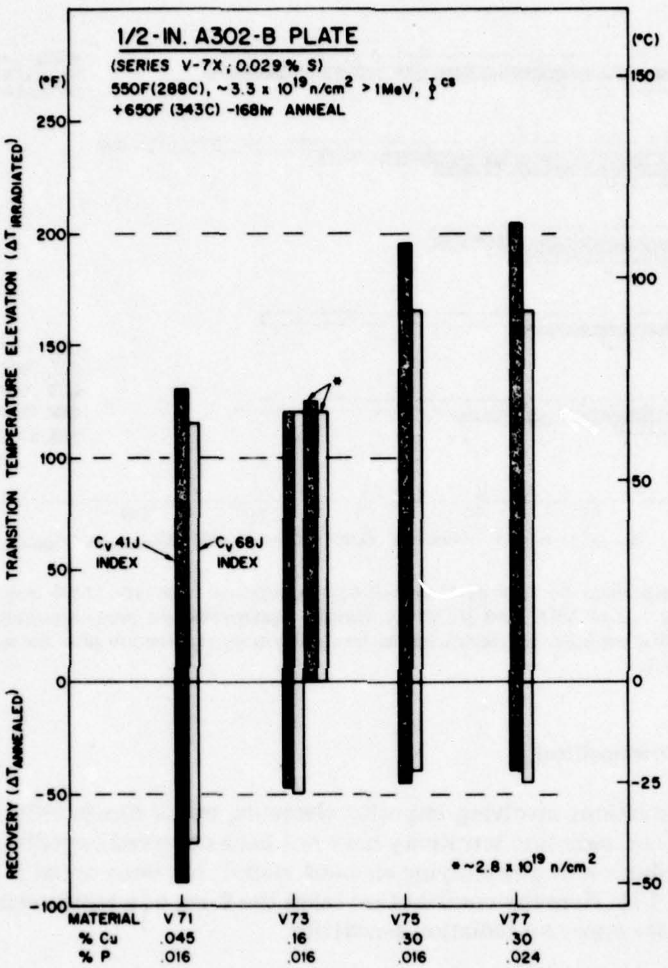

Fig. 10 — Summary of Charpy-V-notch transition-temperature changes (41-J and 68-J indices) of plate series V7X with 288°C (550°F) irradiation and with 343°C (650°F) postirradiation heat treatment. (From Ref. 14.)

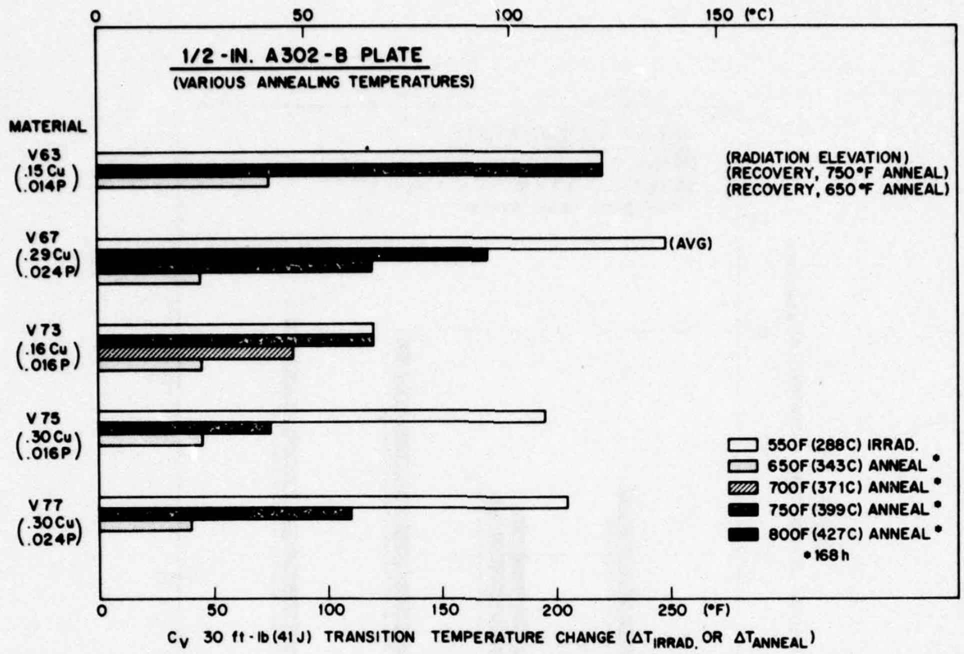

Fig. 11 — Comparison of Charpy-V transition-temperature recovery (41-J index) produced in selected plates (series V6X and V7X) by various postirradiation heat-treatment temperatures. The initial radiation-induced elevations in transition temperature are also shown for reference. (From Ref. 14.)

Alloy Element Composition

Unlike observations involving impurity elements, major direct influences of specific alloying elements on radiation sensitivity have not been observed experimentally. However, an indirect contribution of one alloying element, nickel, has been noted in weld deposit assessments [13, 17]. Here the contribution takes the form of a reinforcement of the detrimental effect of copper on radiation sensitivity.

Data to test the general effects of alloying-element variations in A302-B, A533-B, and A508-2 steels and companion weld metals (submerged arc, manual metal arc deposits) are not currently available. Clarification of the role of certain elements including manganese, chromium, and molybdenum as well as nickel would appear to be in order in view of unexplainable differences noted among typical materials.

Product Form

The significance of product form (plate, weld, or forging) to postirradiation heat-treatment response has not been tested directly. Further, a judgment on this variable cannot be made from the data compiled in this report.

Heat-Treatment Temperature

It has been observed that the heat-treatment temperature must exceed the prior irradiation temperature to obtain measurable transition-temperature recovery or recovery of upper-shelf energy. For 288°C irradiation conditions it has also been demonstrated that notch ductility recovery increases with increasing temperature above 288°C but not linearly (Figs. 1 and 11). Significantly, full upper-shelf recovery can be obtained at a lower temperature than full transition-temperature recovery. For example, 343°C annealing produced full upper-shelf recovery for all but one of the plates of the A302-B melt series (Table 1) but only small transition-temperature recovery [14]. Accordingly the kinetics of upper-shelf recovery appear to be different from that of transition-temperature recovery. Finally, as demonstrated in Figs. 1 and 11, a heat treatment of 427°C following 288°C irradiation may not be sufficiently high in temperature to produce full transition-temperature recovery.

Heat-Treatment Duration

Sufficient data exist to show that a heat-treatment period of 168 hr (1 week) is about optimum for the annealing of plate and weld-deposit materials following 288°C irradiation. Experimental results in support of this assessment are shown in Figs. 1, 12, and 13. For heat-treatment periods shorter than 168 hr, an increase in annealing time generally results in increased recovery (Fig. 1). Extension of the heat-treatment period beyond 168 hr, on the other hand, does not produce a significant increase in either transition-temperature recovery or upper-shelf recovery (Figs. 12 and 13) [4]. The data of Fig. 5 would appear to be in conflict with the preceding conclusion on optimum annealing time; however, the irradiation temperature employed was only 138°C (280°F).

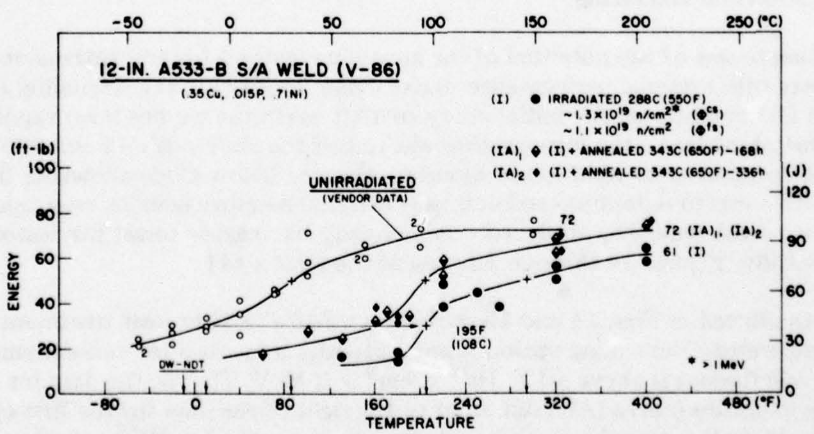

Fig. 12 — Notch ductility recovery of weld V86 by 343°C (650°F) annealing heat treatment for two different times following first-cycle irradiation. (From Ref. 4.)

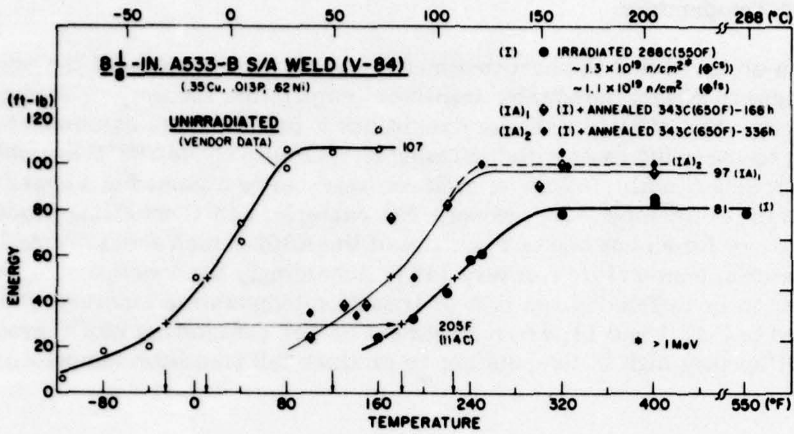

Fig. 13 — Notch ductility recovery of weld V84 by 343°C (650°F) annealing heat treatment for two different times following first-cycle irradiation. (From Ref. 4.)

A 168-hr heat treatment undoubtedly will represent only a small fraction of the total time involved in preparing a reactor system for annealing and in bringing the reactor vessel to temperature. Accordingly a heat-treatment duration of 168 hr is not considered to be overly long from an operations standpoint. Equally important the possibility for falling short of a recovery target becomes significantly less with a 168-hr anneal than with a much shorter heat treatment.

Cyclic Irradiation and Annealing

The ultimate test of the potential of the annealing method for embrittlement relief obviously rests with material performance under cyclic irradiation (I), annealing (A), and reirradiation (R) conditions. An initial study of IAR performance has been reported by NRL [4]. One objective of the investigation was to test the ability of 343 and 399°C intermediate heat treatments to hold notch ductility changes below Code-allowable limits. A second objective was to determine and compare material reembrittlement rates upon reirradiation. Two weld deposits, produced commercially by reactor vessel fabricators, were used for the study. Figures 14 through 18 present the results [4].

The data plotted in Figs. 14 and 15 show that a 343°C 168-hr heat treatment depicting the "low temperature" annealing option is not a promising method for embrittlement relief if the first-cycle fluence is above $\approx 1 \times 10^{19}$ n/cm^2 > 1 MeV. That is, the data for the second-cycle exposure (curve IAR) fall on or to the right of the data for the first-cycle exposure (curve I). In this case the second-cycle fluence was only 3×10^{18} n/cm^2 > 1 MeV. In contrast (Figs. 16 and 17) a 399°C 168-hr heat treatment representing the "high temperature" option proved to be quite effective in reducing the buildup of radiation effects. Transition-temperature changes observed with IAR are summarized in Fig. 18. The data trends clearly show that the rate of reembrittlement after annealing is greater than the rate

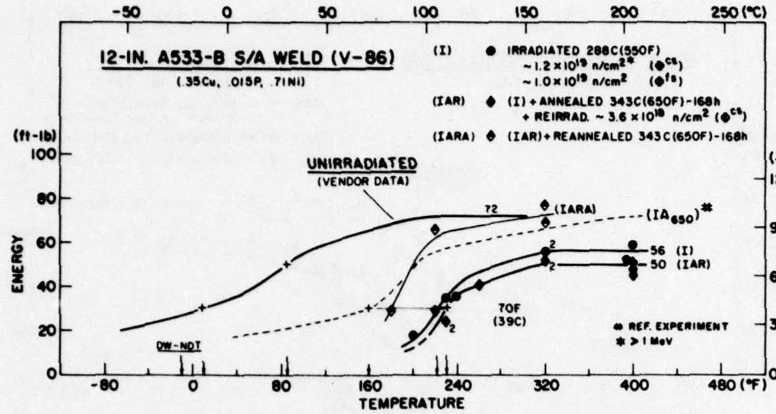

Fig. 14 — Notch ductility behavior of weld V86 after reirradiation following a midcycle 343°C (650°F) annealing heat treatment (curve IAR) and after reirradiation and reheat treatment at 343°C (650°F) (curve IARA). The notch ductility of the weld after the first-cycle irradiation (curve I) and after the first-cycle heat treatment (curve IA, reference experiment) are also shown. (From Ref. 4.)

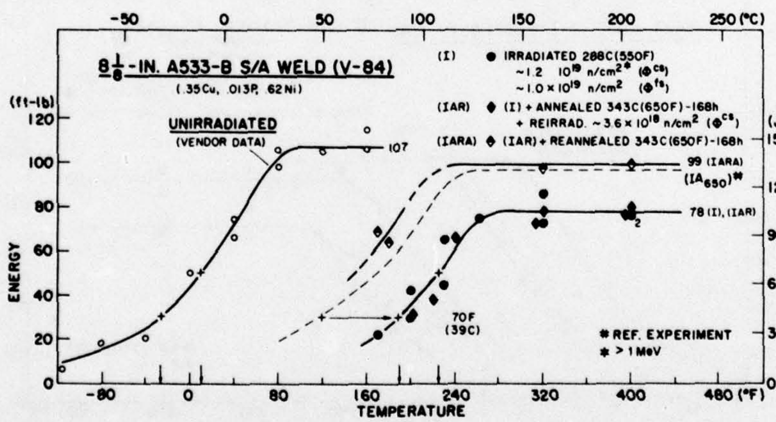

Fig. 15 — Notch ductility behavior of weld V84 after reirradiation following midcycle 343°C (650°F) annealing heat treatment (curve LAR) and after reirradiation and reheat treatment at 343°C (650°F) (curve LARA). The notch ductility of the weld after the first-cycle irradiation (curve I) and after the first-cycle heat treatment (curve IA, reference experiment) are also shown. (From Ref. 4.)

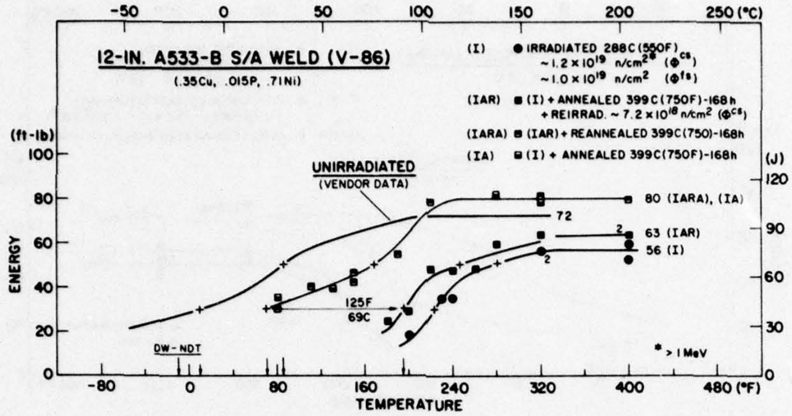

Fig. 16 — Notch ductility behavior of weld V86 after reirradiation following a midcycle 399° C (750° F) annealing heat treatment (curve IAR) and after reirradiation and reheat treatment at 399° C (750° F) (curve IARA). The notch ductility of the weld after the first-cycle irradiation (curve I) and after the first-cycle heat treatment (data points IA) are also shown. (From Ref. 4.)

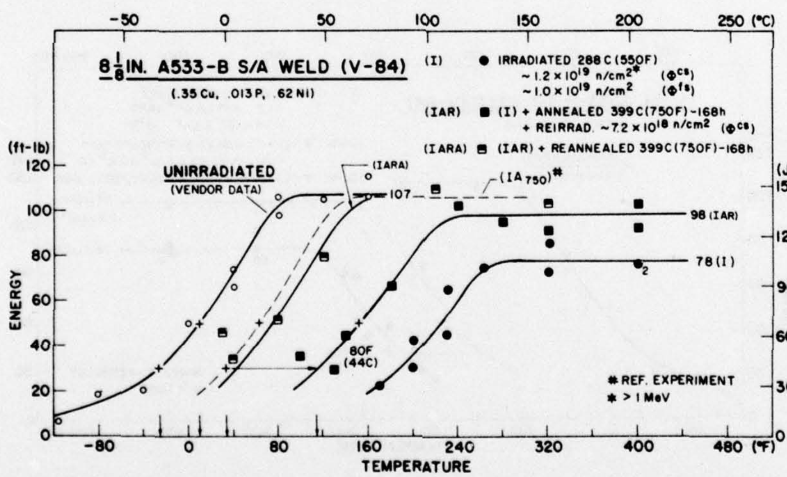

Fig. 17 — Notch ductility behavior of weld V84 after reirradiation following a midcycle 399°C (750°F) annealing heat treatment (curve IAR) and after reirradiation and reheat treatment at 399°C (750°F) (curve IARA). The notch ductility of the weld after the first-cycle irradiation (curve I) and after the first-cycle heat treatment (curve IA) are also shown for reference. (From Ref. 4.)

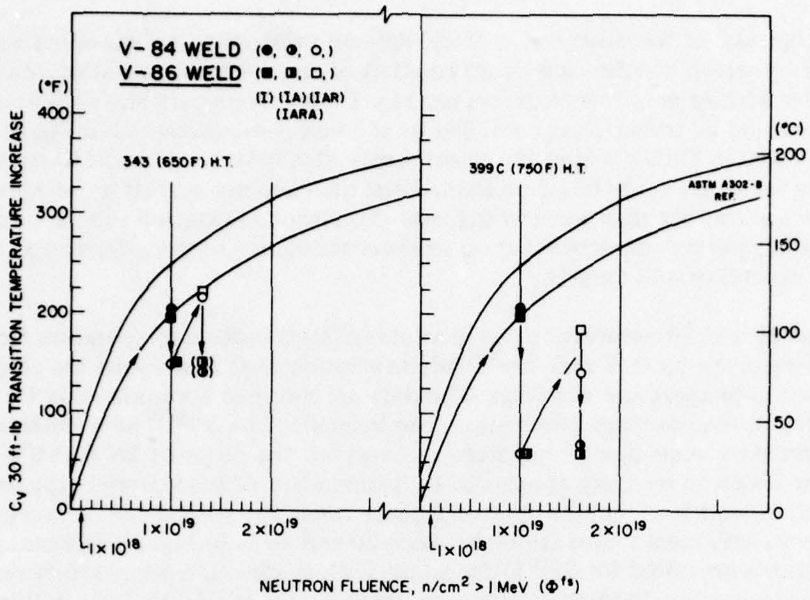

Fig. 18 — Transition-temperature behavior of welds V84 and V86 with 288°C (550°F) irradiation and 343°C (650°F) annealing and with 288°C (550°F) irradiation and 399°C (750°F) annealing (two cycles each). The shaded band refers to the transition-temperature trend of the ASTM A302-B reference plate with < 232°C (450°F) irradiation. (From Ref. 4.)

of embrittlement experienced by nonannealed material. Moreover the trends suggest that that "damage" most readily introduced into the material (that produced early in radiation service) is also that "damage" most readily removed by annealing. This projection is based in part on the similarity of the radiation embrittlement rates observed for the annealed material and the virgin material.

Important insight into IAR performance has been secured with the preceding results; however, additional tests of IAR trends are necessary. At the same time, it must be recognized that reactor experiment operations required by IAR studies are time consuming and much more complex than I or IA experiment operations. Thus, a large volume of data of this type cannot be expected soon. One question which needs an early answer is whether 343°C annealing has potential for reactor vessels having first-cycle fluences less than 1×10^{19} n/cm^2. Also, Figs. 14 and 15 show that upper-shelf changes with annealing cannot be predicted closely from transition-temperature changes. Accordingly upper-shelf IAR patterns must be established separately.

SURVEY OF EXPERIMENTAL ANNEALING RESULTS

The objective of the survey of available data on postirradiation annealing was to reveal trends by which clarification or affirmation of the foregoing observations could be made and by which the influence of certain, as yet unresolved, variables such as composition effects could be tested. Data compiled by the survey are presented in Figs. 19 through 22 and are listed in Tables 2a and 2b. These survey data must be interpreted with caution, because the values are often based on limited test data and not on full-transition curves. Also, some uncertainty may exist in material chemical composition (Table 3) or initial (preirradiation) properties, depending on whether the source of the information is testing-laboratory reports or mill reports.

In Figs 19 and 20 observations on absolute (Δ°C) transition-temperature recovery and percentage recovery by 343 and 399°C postirradiation heat treatments are compared to prior transition-temperature elevation. The data are grouped approximately by neutron fluence level. Several primary observations can be made: The 343°C heat treatment consistently produced a transition-temperature recovery on the order of 28°C (50°F) ± 16°C (30°F). The observed recovery appears to be independent of the material type evaluated, its prior embrittlement level, and its neutron fluence level. The percentage recovery also shows a consistency, with most values falling between 20 and 40%. In Fig. 20 different relationships and trends are noted for 399°C annealing. Higher transition-temperature recoveries are illustrated, as expected. Percentage recovery typically is about 55 to 75%. In this case a trend toward higher recovery with increasing fluence is suggested by the data. A greater spread in values (±22°C (±40°F)) is also shown, which could be indicating a composition effect or an effect of prior transition-temperature elevation. On the other hand, specific contributions of these variables are not evident from the data patterns.

Figures 21 and 22 compare upper-shelf recovery by annealing against the prior upper-shelf reduction by irradiation. For 343°C annealing (Fig. 21) a wide variation in percentage recovery is observed, even though the percentage reductions by irradiation predominantly are in the range of 10 to 30%. Likewise the absolute recovery in upper-shelf energy does not show a particular dependence on the absolute reduction by irradiation. It is suspected that material variables are contributing to the observed differences in annealing response. Neutron fluence level, on the other hand, does not appear to be a critical variable. With 399°C annealing (Fig. 22) essentially all of the materials showed full upper-shelf recovery. The exceptions to this trend, two high fluence points, may be due to the limited number of specimen tests performed. The influence of the prior upper-shelf reduction on subsequent recovery cannot be assessed in Fig. 22 because of the similarity of the radiation changes.

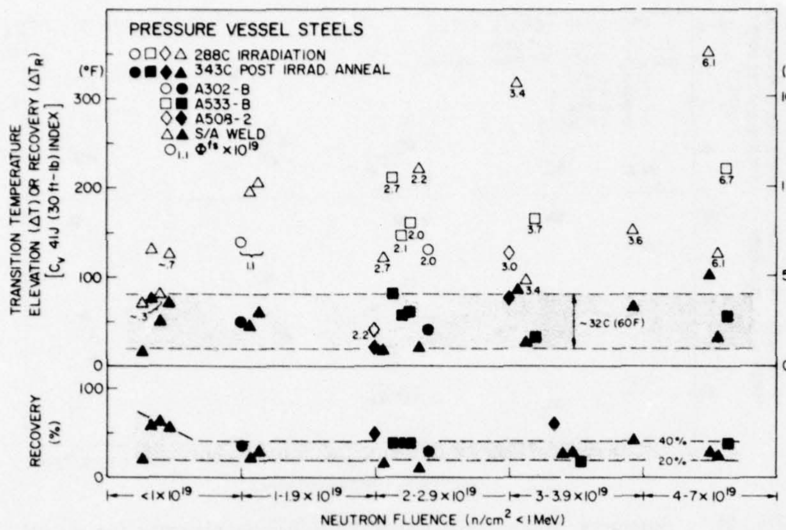

Fig. 19 — Summary of Charpy-V transition-temperature (41-J index) changes observed for several steels with 288°C (550°F) irradiation (open symbols) and with 343°C (650°F) postirradiation heat treatment (filled symbols). The shaded bands illustrate the general recovery trends with increasing fluence.

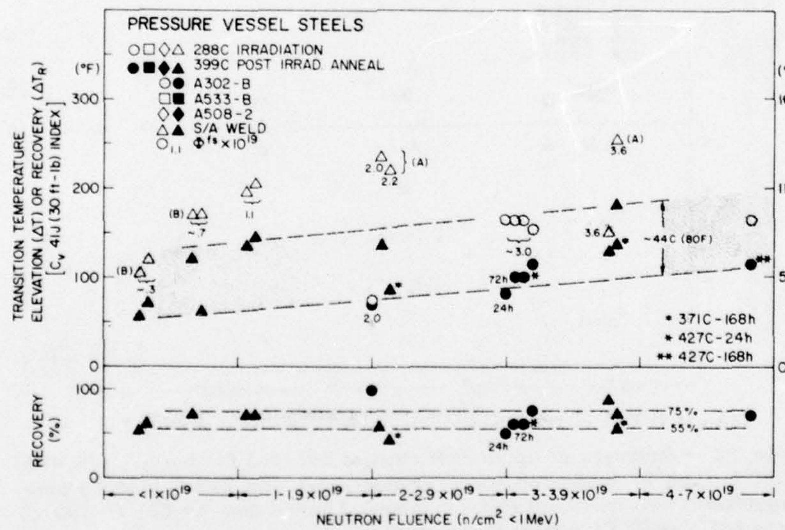

Fig. 20 — Summary of transition-temperature changes (41-J index) observed for several steels with 288°C (550°F) irradiation (open symbols) and with 399°C (750°F) postirradiation heat treatment (filled symbols). Limited data for 371°C (700°F) and 427°C (800°F) annealing are also shown.

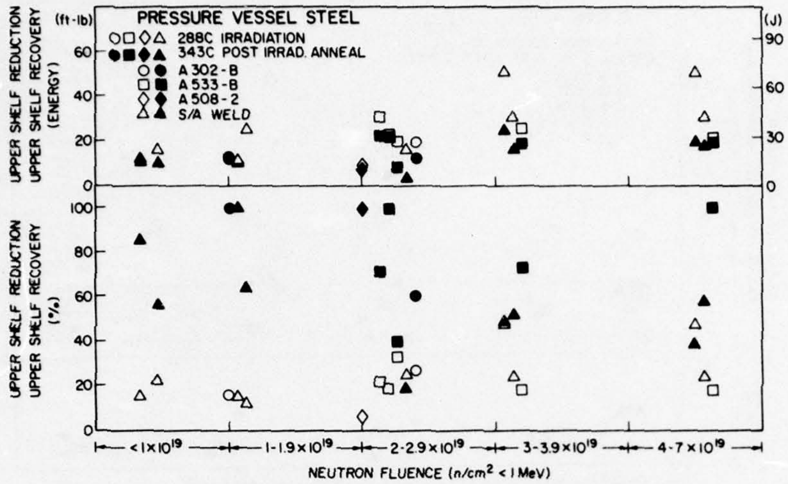

Fig. 21 — Summary of Charpy-V upper-shelf changes observed for several steels with 288°C (550°F) irradiation (open symbols) and with 343°C (650°F) postirradiation heat treatment (filled symbols)

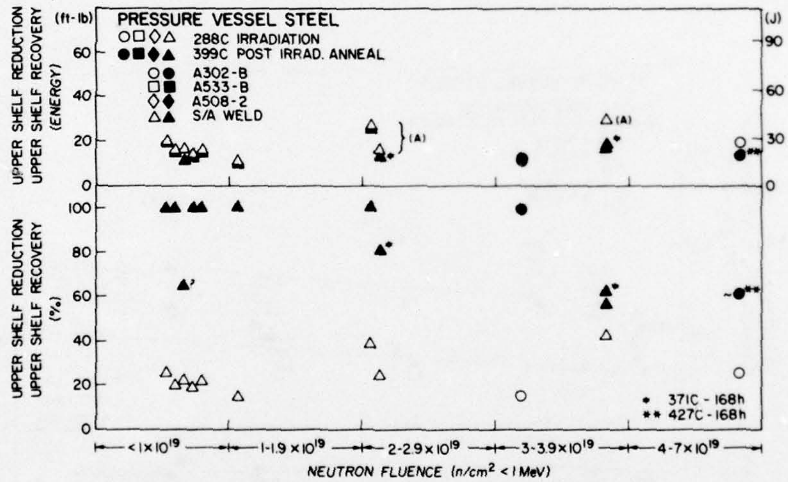

Fig. 22 — Summary of upper-shelf changes observed for several steels with 288°C (550°F) irradiation (open symbols) and with 399°C (750°F) post-irradiation heat treatment (filled symbols). Limited data for 371°C (700°F) and 427°C (800°F) annealing are also shown.

Table 2a — Data Compilation (Metric Units) for Notch Ductility Recovery by Postirradiation Heat Treatment

Code No.	Upper-Shelf Energy (J)	Fluence (10^{19} n/cm^2 > 1 MeV)	Irradiation Effects: ΔT_{41} (°C)	Irradiation Effects: ΔT_{68} (°C)	Irradiation Effects: ΔUS (J)	Anneal (°C-hr)	Annealing Recovery: ΔT_{41} (°C)	Annealing Recovery: ΔT_{41} (%)	Annealing Recovery: ΔT_{68} (°C)	Annealing Recovery: ΔUS (J)	US (%)	Experiment Number
A302-B (Plate) (ASTM Reference)												
D36	118	3.0	92	89	43	399-24	44	49	44	≈ 7	≈ 16	L[a](C18)13
						399-72	56	61	53	—[b]	—	L(C18)13
						427-24	56	61	—	—	—	L(C18)13
F23	111	1.1	78	81	18	343-168	28	36	17	18	100	L(C57)88
F26	98	3.1	86	78	12	399-168	≈ 64	≈ 74	≈ 42	≈ 7	≈ 56	L(C55)54
F23	106	7.1 (307°C)[c]	92	83	27	427-168	64	70	61	16	60	BRPR[d]-1 (122-3)
D36	118	1.4 (254°C)	111	111	49	316-24	31	28	39	≈ 8	≈ 17	L(C55)31
F23	111	1.0 (260°C)	100	92	22	343-168	50	50	28	≈22	≈100	L(C57)88
D36	118	3.0 (338°C)	61	58	39	427-24	39	64	33	—	—	L(C18)13
F26	98	1.7 (371°C)	17	22	12	454-168	≈ 17	≈100	≈ 22	≈12	≈100	L(C18)42
A302-B (Plate)												
MBU	172	2.0	42	56	29	399-168	42	100	56	—	—	T[e](C3)58
40C	102	2.0	72	83	27	343-168	≈ 22	≈ 31	≈ 31	16	60	B[f](B4)-2
						371-168	—	—	—	19	70	B(B4)-2
A533-B (Plate)												
1T	81	2.0	89	—	27	343-168	≈ 60	≈ 38	—	11	18	B(B4)-2
						371-168	—	—	—	27	100	B(B4)-2
Q98	189	2.7	117	114	42	343-168	44	38	33	30	71	T(D3)74
						343-336	44	38	—	—	—	T(D3)74
CEP1	156	2.1	81	103	30	343-168	31	38	33	30	100	T(D3)75
		6.7	117	108	29	343-168	28	24	17	27	≈100	T(B3)76
CEP2	190	3.7	92	92	35	343-168	17	18	11	26	73	T(D3)75
N27	186	2.5	78	86	24	343-168	—	—	≈ 50	24	100	B(B4)-2
						371-168	—	—	78	27	100	B(B4)-2
N27	186	2.3 (260°C)	81	92	34	343-168	≈ 36	≈ 45	53	—	—	T(A4)64
N27	186	2.6 (227°C)	167	153	45	343-168	≈122	≈ 73	≈106	—	—	T(A4)64
N29	196	2.3 (260°C)	28	28	12	343-168	< 11	< 40	< 11	—	—	T(A4)64
N29	196	2.6 (227°C)	103	114	34	343-168	≈ 78	≈ 76	89	—	—	T(A4)64
A533-B (Weld)												
Q96	210	2.7	61	72	61	343-168	< 11	< 18	11	≈30	≈ 49	T(D3)74
CEW1	145	3.4	175	189	69	343-168	47	27	56	34	49	T(D3)75
		6.1	194	208	69	343-168	56	29	56	27	39	T(B3)76
CEW2	175	3.4	53	75	42	343-168	14	26	13	22	≈ 52	T(D3)75
		6.1	69	97	42	343-168	≈ 17	≈ 24	≈ 39	≈24	≈ 58	T(D3)76
W	88	2.2	122	—	22	343-168	11	9	≈ 17	4	19	B(B4)-2
						371-168	47	39	≈ 39	18	81	B(B4)-2
W	94	2.0	131	—	37	399-168	75	57	—	37	100	B(B4)-6
W	94	≈3.6	142	—	39	399-168	100	71	—	≈23	≈ 59	T(D3)78
						371-168	≈ 75	≈ 53	—	≈11	≈ 63	T(D3)78
N8	78	≈0.3	72	69	16	343-168	42	58	—	≈14	≈100	B(C2)17
		≈0.7	100	92	27	399-168	—	—	—	≈30	≈100	B(C2)20
AW	163	≈3.6	≈ 83	≈ 92	24	399-168	72	87	81	—	—	T(D3)78
						343-168	36	43	36	—	—	T(D3)78
N6	107	1.3	111	100	30	343-168	—	—	—	11	100	B(C2)14
V84	144	1.1	114	119	34	343-168	33	29	31	22	64	B(B4)10
						343-336	33	29	31	26	76	B(B4)10
						399-168	51	71	89	34	100	B(B4)10
V86	98	1.1	108	117	15	343-168	25	23	50	15	100	B(B4)10
						343-336	25	23	50	15	100	B(B4)10
						399-168	75	69	89	24	100	B(B4)10
N1	107	≈0.3	42	78	4	399-168	≈ 42	≈100	78	—	—	B(C2)17
N2	104	≈0.3	58	56	14	399-168	31	52	33	81	—	B(C2)17
		≈0.7	94	89	23	399-168	67	71	—	≈15	> 65	B(C2)20
N3	100	≈0.3	67	67	4	399-168	≈ 39	≈ 58	—	—	—	B(C2)17
		≈0.7	94	94	19	399-168	33	35	—	≈31	≈100	B(C2)20
N4	98	≈0.3	44	28	8	343-168	28	63	8	41	—	B(C2)17
		≈0.7	69	58	22	343-168	—	—	—	≈12	≈ 56	B(C2)20
						399-168	—	—	—	22	100	B(C2)20
N7	111	≈0.3	39	42	5	343-168	≈ 8	≈ 21	42	—	—	B(C2)17
		≈0.7	69	72	22	399-168	—	—	—	35	100	B(C2)20
A508-2 (Forging)												
Q41	218	2.2	22	25	12	343-168	≈ 11	≈ 50	11	≈12	100	T(D3)72
Q71	186	3.0	69	83	29	343-168	42	60	64	—	—	T(D3)72

[a]Low Intensity Test Reactor (LITR).
[b]Not determined.
[c]307°C irradiation.
[d]Big Rock Point Reactor (BRPR).
[e]*Union Carbide Research Reactor* (UCRR).
[f]SUNY at Buffalo Reactor (UBR).

Table 2b — Data Compilation (English Units) for Notch Ductility Recovery by Postirradiation Heat Treatment

Code No.	Upper Shelf Energy (ft-lb)	Fluence (10^{19} n/cm^2 > 1 MeV)	Irradiation Effects			Anneal	Annealing Recovery				US (%)	Experiment Number
			ΔT_{30} (°F)	ΔT_{50} (°F)	ΔUS (ft-lb)	(°F-hr)	ΔT_{30} (°F)	ΔT_{30} (%)	ΔT_{50} (°F)	ΔUS (ft-lb)		
A302-B (Plate) (ASTM Reference)												
D36	87	3.0	165	160	32	750-24	80	49	80	≈ 5	≈ 16	L[a](C18)13
						750-72	100	61	95	—[b]	—	L(C18)13
						800-24	100	61	—	—	—	L(C18)13
F23	82	1.1	140	145	13	650-168	50	36	30	13	100	L(C57)88
F26	72	3.1	155	140	9	750-168	≈115	≈ 74	≈ 75	≈ 5	≈ 56	L(C55)54
F23	78	7.1 (585°F)[c]	165	150	20	800-168	115	70	110	≈12	≈ 60	BRPR[d]-1 (122-3)
D36	87	1.4 (490°F)	200	200	36	600-24	55	28	70	≈ 6	≈ 17	L(C55)31
F23	82	1.0 (500°F)	180	165	16	650-168	90	50	50	≈16	≈100	L(C57)88
D36	87	3.0 (640°F)	110	105	29	800-24	70	64	60	—	—	L(C18)13
D36	87	3.0 (740°F)	65	65	25	900-24	≈ 55	≈ 85	—	—	—	L(C18)13
F26	72	1.7 (700°F)	30	40	9	850-168	≈ 30	≈100	≈ 40	≈ 9	≈100	L(C18)42
A302-B (Plate)												
MBU	127	2.0	75	100	21	750-168	75	100	100	—	—	T[e](C3)55
40C	75	2.0	130	150	20	650-168	≈ 40	≈ 31	≈ 55	12	60	B[f](B4)-2
						700-168	—	—	—	14	70	B(B4)-2
A533-B (Plate)												
1T	60	2.0	160	—	20	650-168	≈ 60	≈ 38	—	8	18	B(B4)-2
						700-168	—	—	—	20	100	B(B4)-2
Q98	139	2.7	205	210	31	650-168	80	38	60	22	71	T(D3)74
						650-336	80	38	—	—	—	T(D3)74
CEP1	115	2.1	145	185	22	650-168	55	38	60	22	≈100	T(D3)75
		6.7	210	195	21	650-168	50	24	30	20	100	T(B3)76
CEP2	110	3.7	165	165	26	650-168	30	18	20	19	73	T(D3)75
N27	137	2.5	≈140	155	18	650-168	—	—	≈ 90	18	100	B(B4)-2
						700-168	—	—	140	20	100	B(B4)-2
N27	137	2.3 (500°F)	145	165	25	650-168	≈ 65	≈ 45	95	—	—	T(A4)64
N27	137	2.6 (440°F)	300	275	33	650-168	≈220	≈ 73	≈190	—	—	T(A4)64
N29	145	2.3 (500°F)	50	50	9	650-168	⩽ 20	< 40	⩽ 20	—	—	T(A4)64
N29	145	2.6 (440°F)	185	205	25	650-168	≈140	≈ 76	160	—	—	T(A4)64
A533-B (Weld)												
Q96	155	2.7	110	130	45	650-168	< 20	⩽ 18	20	22	49	T(D3)74
CEW1	107	3.4	315	340	51	650-168	85	27	100	25	49	T(D3)75
		6.1	350	375	51	650-168	100	29	100	20	39	T(B3)76
CEW2	129	3.4	95	135	31	650-168	25	26	23	16	52	T(D3)75
		6.1	125	175	31	650-168	≈ 30	≈ 24	≈ 70	≈18	≈ 58	T(B3)76
W	65	2.2	220	—	16	650-168	20	9	≈ 30	3	19	B(B4)-2
						700-168	85	39	≈ 70	13	81	B(B4)-2
W	69	2.0	235	—	27	750-168	135	57	—	27	100	B(B4)-6
W	69	3.6	255	—	29	750-168	180	71	—	≈17	≈ 59	T(D3)-78
						700-168	≈135	≈ 53	—	≈19	≈ 63	T(D3)-78
N8	78	≈0.3	130	125	12	650-168	75	58	—	≈10	≈100	B(C2)17
		≈0.7	180	165	20	750-168	—	—	—	≈22	≈100	B(C2)20
AW	120	≈3.6	≈150	≈165	18	650-168	65	43	65	—	—	T(D3)78
						750-168	130	87	145	—	—	T(D3)78
N6	79	1.3	200	180	22	650-168	—	—	22	100	—	B(C2)14
V84	106	1.1	205	215	25	650-168	60	29	55	16	64	B(B4)10
						650-336	60	29	55	19	76	B(B4)10
						750-168	145	71	160	25	100	B(B4)10
V86	72	1.1	195	210	11	650-168	45	23	90	11	100	B(B4)10
						650-336	45	23	90	11	100	B(B4)10
						750-168	135	69	160	18	100	B(B4)10
N1	79	≈0.3	75	140	3	750-168	≈ 75	≈100	140	—	—	B(C2)17
N2	77	≈0.3	105	100	10	750-168	55	52	60	60	—	B(C2)17
		≈0.7	170	160	17	750-168	120	71	—	≈11	> 65	B(C2)20
N3	74	≈0.3	120	120	3	750-168	≈ 70	≈ 58	—	—		B(C2)17
		≈0.7	170	170	14	750-168	60	35	—	≈23	≈100	B(C2)20
N4	72	≈0.3	80	50	6	650-168	50	63	15	30	—	B(C2)17
		≈0.7	125	105	16	650-168	—	—	—	≈ 9	≈ 56	B(C2)20
						750-168	—	—	—	16	100	B(C2)20
N7	82	≈0.3	70	75	4	650-168	≈ 15	≈ 21	75	—	—	B(C2)17
		≈0.7	1[illegible]5	130	16	750-168	—	—	—	26	100	B(C2)20
A508-2 (Forging)												
Q41	161	2.2	40	45	9	650-168	≈ 20	≈ 50	20	≈ 9	100	T(D3)72
Q71	137	3.0	125	150	21	650-168	75	60	115	—	--	T(D3)72

[a]Low Intensity Test Reactor (LITR).
[b]Not determined.
[c]585°C irradiation.
[d]Big Rock Point Reactor (BRPR).
[e]Union Carbide Research Reactor (UCRR).
[f]SUNY at Buffalo Reactor (UBR).

Table 3 — Chemical Compositions of Survey Materials (Plate, Forging, Weld Deposit)

MATERIAL	CODE	Chemical Composition (wt-%) Cu	P	C	Mn	S	Si	Ni	Cr	Mo
A302-B	D36	0.20	0.011	0.24	1.34	0.023	0.23	0.18	0.11	0.51
(Plate)	F23	0.20	0.011	0.24	1.34	0.023	0.23	0.18	0.11	0.51
(ASTM Ref)	F26	0.20	0.011	0.24	1.34	0.023	0.23	0.18	0.11	0.51
A302-B	MBU	0.15	0.006	0.25	1.37	0.011	0.21	0.26	0.11	0.63
(Plate)	40C	0.008	0.021	0.23	1.30	0.022	0.21	—	—	0.53
A533-B	1T	0.24	0.012	0.22	1.30	0.024	0.28	0.52	—	0.40
(Plate)	Q98	0.22	0.006	0.25	1.44	0.010	0.23	0.58	0.12	0.52
	CEP1	0.17	0.009	0.23	1.29	0.015	0.21	0.56	0.10	0.57
	CEP2	0.24	0.008	0.25	1.40	0.011	0.23	0.62	0.11	0.59
	N27	0.13	0.008	0.17	1.22	0.008	0.19	0.58	—	0.50
	N29	0.03	0.008	0.17	1.21	0.007	0.20	0.56	—	0.50
A533-B*	Q96	0.26	0.010	0.17	1.17	0.010	0.18	0.18	0.08	0.51
(Weld)	CEW1	0.36	0.015	0.14	1.35	0.012	0.22	0.78	0.07	0.55
	CEW2	0.20	0.016	0.13	1.11	0.013	0.17	0.04	0.05	0.55
	W	0.35	0.020	0.09	1.45	0.013	0.68	0.57	—	0.39
	AW	0.055	0.022	0.11	1.33	0.012	0.44	0.97	0.07	0.54
	V84	0.35	0.013	0.14	1.56	0.011	0.14	0.62	0.03	0.53
	V86	0.35	0.016	0.08	1.60	0.013	0.55	0.69	—	0.40
A508-2	Q41	0.09	0.005	0.21	0.68	0.008	0.27	0.71	0.39	0.59
(Forging)	Q71	0.13	0.007	0.19	0.69	0.009	0.31	0.82	0.38	0.62

*Chemical compositions of welds N1 to N8 are not available.

SUMMARY

Reactor pressure-vessel steels including A302-B, A533-B, A508-2, and companion weld metals exhibit significant variability in their response to postirradiation heat treatment for embrittlement relief. Also, a difference in response to postirradiation heat treatment has been observed between transition-temperature and upper-shelf-energy C_v properties for a given material.

Several metallurgical and service factors are suspected of contributing to the variation in notch ductility recovery and are identified. Within the limitations of the existing experimental data, the contributions or importance of certain of these factors could be assessed using direct or indirect test comparisons. Table 4 identifies where assessments were permitted by the data and the qualification made in each case. Among those variables identified as contributing factors, annealing temperature and duration are ranked as having the greatest influence on the recovery of properties. This relationship is in turn reflected in the importance of heat-treatment time and conditions selected for cyclic annealing and reirradiation treatments. Impurity-element composition (copper content) is ranked second in importance. Here a detrimental effect of a high copper content (0.30% Cu) on upper-shelf recovery was demonstrated for A302-B steel. A similar effect for companion weld deposits is quite probable. Of those factors yet to be qualified, four should be given first priority. The significance of relative radiation resistance and of alloying-element composition should be established to preclude possible technical surprises in future annealing operations. Annealing recovery with different first-cycle fluences and second-cycle fluences (repeat irradiation) should be evaluated to help establish optimum heat treatment schedules which provide maximum benefit and flexibility.

The present data show that 343°C annealing does not have high promise for those reactor applications having first-cycle fluences exceeding 1×10^{19} n/cm^2 > 1 MeV. On the other hand, it is clear that 399°C annealing or annealing at somewhat higher temperatures does have significant promise for the embrittlement relief of reactor vessels. With this heat treatment, transition-temperature recoveries of about 55% minimum and 75% maximum can be expected for most materials, based on the data survey. More importantly, full upper-shelf recovery for fluences up to 3×10^{19} n/cm^2 appears typical for the materials of concern.

Although major progress has been made, the present state of the art is insufficiently advanced to permit an immediate consideration of the annealing method for the relief of reactor vessel embrittlement. One basis for this opinion is that experimental data describing property changes with reirradiation are too sparse to confidently base engineering judgments and property projections for this condition. An equally important basis is that knowledge of factors governing reirradiation behavior (sensitivity) is extremely limited. To compound both issues, it has been established that the rate of reembrittlement of annealed material is significantly different (greater) from that of nonannealed material at an equivalent fluence level. The trend in upper-shelf reduction with reirradiation likewise appears to be significantly different from that of nonannealed material. Beyond first-cycle behavior, information on notch ductility trends with multiple annealing and reirradiation cycles is in comparison practically nonexistent. Thus, although high-temperature annealing shows promise for embrittlement relief, further research must precede its application. Specifically, further research is needed on single-cycle and multicycle annealing and reirradiation treatments. For maximum

Table 4 — Status of Experimental Assessments of Variables Suspected of Contributing to the Alleviation of Radiation Embrittlement by Postirradiation Annealing

Variable Description	Experimental Qualification		
	Contributing	Noncontributing	Not Established
Irradiation temperature	X		
Neutron fluence	X[a]		
Relative radiation resistance			X
Applied Stress:			
During irradiation		X	
During annealing			X
Impurity element composition	X		
Alloying element composition			X
Product form			X
Annealing temperature	X		
Annealing duration	X		
Cyclic anneal conditions (temp., time)	X		
Fluence before first anneal			X
Fluence between anneals			X

[a]Dependent on the heat treatment condition.

usefulness the investigations should concentrate largely on 399°C heat treatments rather than 343°C heat treatments. Concurrently, in-depth studies of the capabilities of the high-temperature annealing option should be verified for the single-cycle irradiation and anneal condition for all projected material, fluence, and service parameters.

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